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Tuning superconductivity by magnetic fields in $Eu(Fe_{0.81}Co_{0.19})_2As_2$

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The distinct difference between BCS-type and unconventional triplet superconductivity manifests itself in their response to external magnetic fields. An applied field easily extinguishes s-wave singlet superconductivity by both the paramagnetic or orbital pair-breaking effects. However, it hardly destroys triplet state because the paramagnetic effect, owing to spins of the Cooper pairs readily aligned with the field, is not so efficacious. This suggests that the triplet superconductivity may be affected mostly by the orbital effect. Conversely, if one can break down the orbital effect then one can recover the superconductivity. Here, we show that superconductivity can be induced with magnetic fields applied parallel to the *ab* plane of crystals of the magnetic $Eu(Fe_{0.81}Co_{0.19})_2As_2$ superconductor. We argue that the tuning superconductivy may be actuated by relative enhancement of ferromagnetic interactions between the Eu^{2+} moments lying in adjacent layers and removal of their canting toward *c* axis that is present in zero field.

$Eu(Fe_{0.81}Co_{0.19})_2As_2$ is one of the members of solid solutions $Eu(Fe_{1-x}Co_x)_2As_2$ crystallizing in the tetragonal $ThCr_2Si_2$-type structure (space group I4/mmm) at room temperature. In the unit cell of the parent $EuFe_2As_2$ compound, the Fe^{2+} ions distributed on the Z = 0.25 layers exhibit a spin-density wave (SDW) ordering below 190 K, while the Eu^{2+} ions located on the Z = 0 layers order antiferromagnetically below 19 K. The magnetic moments of Fe^{2+} and Eu^{2+} are parallel to one another, and are confined to the *ab* plane [1–4]. The suppression of the SDW state in $EuFe_2As_2$, by hydrostatic pressure [5, 6] or by suitable chemical substitution, i.e., $Eu(Fe_{1-x}Co_x)_2As_2$ with $0.18 < x < 0.3$ [7, 8],

results in the emergence of the superconductivity. It was also shown that the occurrence of superconductivity does not significantly change the magnetic ordering temperature T_N of the Eu^{2+} magnetic ions.

ac electrical resistivity ($\rho(T)$) and ac magnetic susceptibility ($\chi'(T)$, $\chi''(T)$) of the $Eu(Fe_{0.81}Co_{0.19})_2As_2$ single crystal, [9] presented in Fig. 1 exemplify the coexistence of magnetism and superconductivity. In the studied compound, the magnetic ordering of the Eu^{2+} ions sets in at the Néel temperature $T_N \sim 16.5$ K and superconductivity at the critical temperature $T_c \sim 5.1$ K.

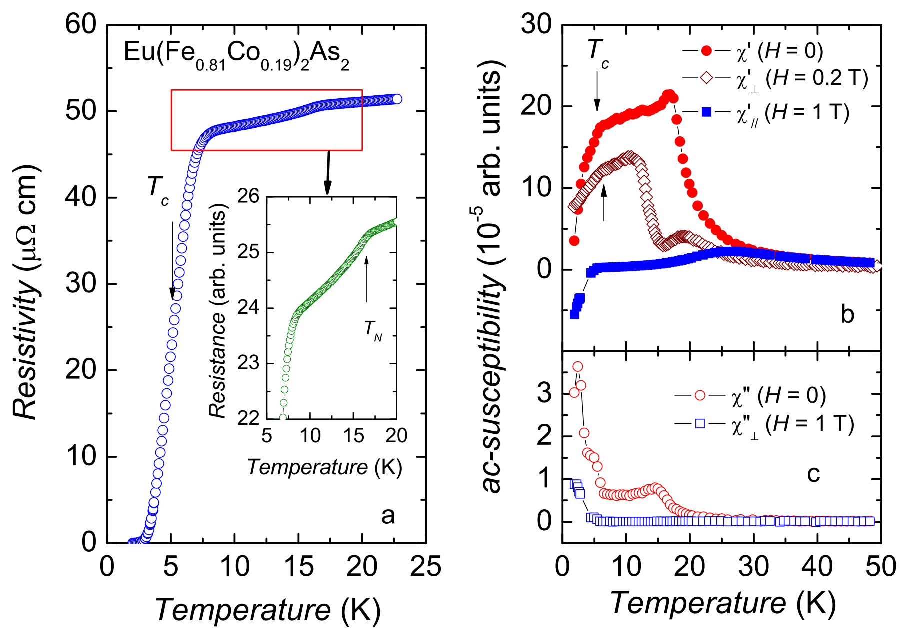

Figure 1: a) Temperature dependence of the ac-electrical resistivity of $Eu(Fe_{0.81}Co_{0.19})_2As_2$ measured with a current of 5 mA. The superconducting transition temperature $T_c \sim 5.1$ K is defined as a midpoint of the resistivity jump. The inset shows a change in the resistivity slope around T_N in an enlarged scale. b) The real component $\chi'(T)$ and c) the imaginary component $\chi''(T)$ of ac-magnetic susceptibility as a function of temperature.

Note that as external field is applied parallel to the *ab* plane the susceptibility reveals two well separated maxima. The low temperature and high temperature anomalies correspond to antiferromagnetic and ferromagnetic components, respectively. For fields above 1 T, $\chi'_{\parallel}(T)$

and $\chi'_{\perp}(T)$ behave similarly, e.g., showing a negative value below T_c and a maximum at $\sim$ 25 K conforming the field-induced ferromagnetic arrangement of Eu^{2+}. We pay attention to the presence of a dissipative process in $\chi''(T)$ below T_N (Fig. 1 c), which is in agreement with the Mössbauer data [10], where the magnetic moments of Eu^{2+} have been designated to be canted from the c axis by an angle of 60°. Because of the magnetic interactions, the Eu^{2+} layers are still expected to be weak conducting layers, and therefore the c axis remains the worse conducting direction than the ab plane.

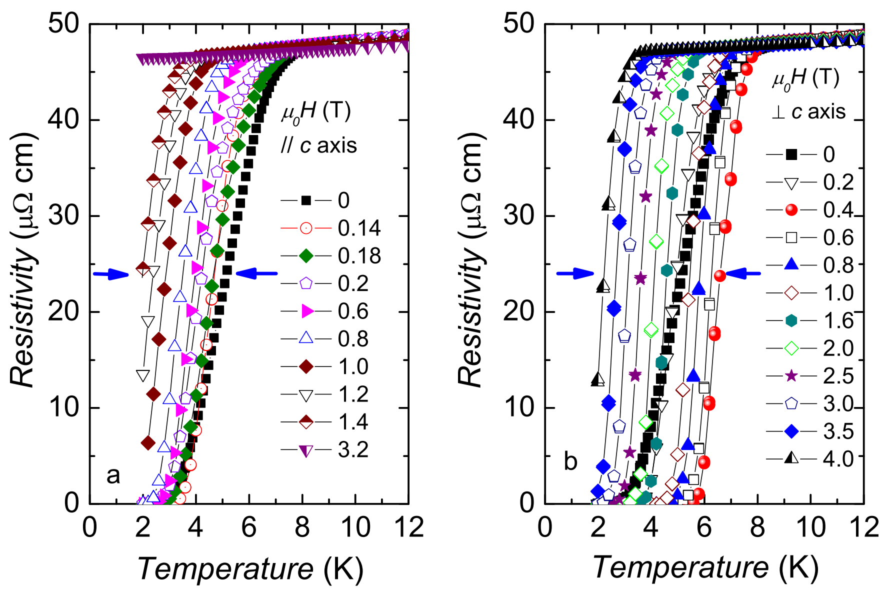

Figure 2: The resistivity as a function of temperature for a) $H\|c$ and b) $H_{\perp}c$. The arrows indicate midpoints of the resistivity curves. A perpendicular field of 0.4 T induces superconductivy by shifting $T_c^{R=0} = 2.2$ K (at 0 T) up to 5.6 K.

The field-dependent resistivity around the superconducting transition is shown in Fig. 2. For $H\|c$, T_c and the superconducting transition width, defined as $\Delta T_c = T90\% - T10\%$, where $T90\%$ and $T10\%$ are the temperatures corresponding to 90% and 10% of the resistivity jump, decrease with increasing field. However, ΔT_c persists with a large value of 1.7 K at fields above 1 T. It is well known that the relative domination of the two pair-breaking effects determines the order of the phase transition in a type-II superconductor. The dominating orbital pair-breaking is usually associated with a second order phase transition while the

dominating paramagnetic pair-breaking is related with a first order phase transition. Owing to a regular dependence of $\Delta T_c(H)$ observed for $H\|c$, no sudden change in the pair-breaking mechanism should be expected.

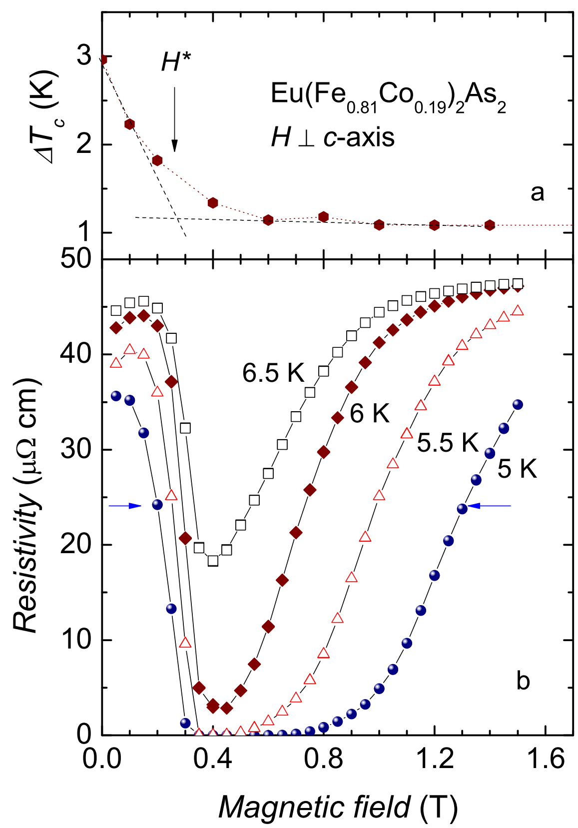

Figure 3: a) The superconducting transition width ΔT_c and b) The electrical resistivity of $Eu(Fe_{0.81}Co_{0.19})_2As_2$ at 5, 5.5, 6 and 6.5 K as a function of $H_{\perp}c$. The dashed lines in a) are linear extrapolations of ΔT_c in low- and high-field regimes. The intersection of these lines corresponds to H^*.

A more salient feature of the resistivity is observed under magnetic fields perpendicular to the c axis (Fig. 2 b). Here we want to point out two experimental facts which indicate important correlations between them. One is a field-induced superconductivity in a wide field range. It is clear seen from the figure that the zero-resistance point $T_c^{R=0}$ as well as T_c shift towards higher temperatures for a range of fields 0.27 - 1 T. The other fact should be noted is that ΔT_c rapidly narrows with increasing magnetic fields, i.e., ΔT_c amounting to

2.96 K at zero field decreases down to 1.34 K at 0.4 T (see Fig. 3 a) and levels off to 1.07 K at fields above 0.6 T. The sharpening transition with increasing fields might be associated with a change in the pair-breaking mechanisms, e.g., from the orbital to dominant paramagnetic one. We believe that the suppression of the orbital pair-breaking effect may explain the field-induced superconductivity in $Eu(Fe_{0.81}Co_{0.19})_2As_2$. Possible other mechanisms accounting for field-induced superconductivity will be discussed below. The field-induced superconductivity is more evident in the isothermal resistivity measurements shown in Fig. 3 b. The normal state of the studied sample in zero field for temperatures between 5 K and 6.5 K can be pushed into a superconducting state via applying a suitable magnetic strength of about $H^* \sim 0.27$ T, i.e., the field strength presumably necessary to abate the orbital pair-breaking effect.

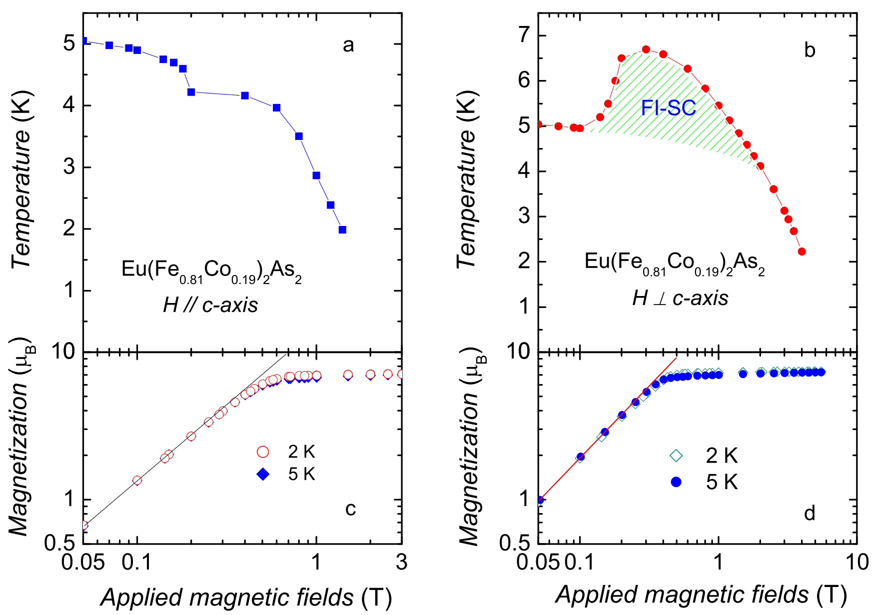

Figure 4: a) and b) The field-temperature phase diagram for $H \| c$ and $H \perp c$, respectively. FI-SC denotes the area of the field-induced superconductivity. c) and d) Magnetization at 2 and 5 K versus $H \| c$ and $H \perp c$, respectively.

The field dependencies of T_c are summarized in temperature versus magnetic field phase diagrams shown in Fig. 4 a and Fig. 4 b. In the vicinity of T_c, one find a linearity between H and T_c , and thus one estimates the initial slope of the $dH/dT|_{T=T_c}$ = -0.33 T/K and

-0.49 T for $H\|c$ and $H \perp c$, respectively. Using the Werthamer-Helfand-Hohenberg formula for a dirty limit[11]:

$$H_{c2}^{orb} = -0.693 T_c \frac{dH_{c2}}{dT}|_{T=T_c} \tag{1}$$

we evaluated the orbital pair-breaking fields $H_{c2}^{orb,\|c} = 1.2$ T and $H_{c2}^{orb,\perp c} = 1.8$ T in absence of any paramagnetic limitation. On the other hand, Clogston has shown that the paramagnetically limited upper critical field should be given by:[12]

$$H_{po} = 1.84 T_c. \tag{2}$$

For $T_c = 5.1$ K and using Eq. 2 we estimated H_{po} to be 9.5 T. An extrapolation of $T_c(H)$ to $T = 0$ yields the value of the upper critical field $H_{c2}(0) \sim 2.7$ T for $H\|c$ and 6.5 T for $H_{\perp}c$. A comparison of these upper critical fields implies that the $T_c(H)$ curve in the $H\|c$ configuration is mainly governed by the orbital pair-breaking effect but a dominating paramagnetic effect is taken down for $H_{\perp}c$. The absence of the orbital limit for high field strength $H_{\perp}c$ is consistent with the behaviour of $\Delta T_c(H)$ considered above.

The dc-magnetization (M) data collected at 2 and 5 K (Fig. 4 c and d) exhibit a change in the slope at about 0.2 T, indicative of spin reorientation towards the magnetic field direction. Above 1 T, corresponding to a ferromagnetic state, $M_{\|}$ and $M_{\perp}$ attain respective value of 7.14 and 7.37 μ_B/f.u, slightly larger than 7 μ_B expected for Eu^{2+}. A possible contribution from polarized Fe^{2+} moments should be checked in future studies. The fact that the superconductivity coexisting with the ferromagnetic ordering over a wide range of magnetic field strongly suggests its unconventional character, presumably of a triplet state with net spin $S = 1$ parallel to ab plane. The other evidences of unconventional superconductivity come from the observation of the field-induced superconductivity and a large anisotropy of $T_c(H)$.

The undubitable difference in the magnetoresistance (MR) for $H\|c$ and $H \perp c$ (Fig. 5) demonstrates a close relationship between the field-induced superconductivity and the disturbance of the antiferromagnetic Eu^{2+} sublattice. The spin-flip process of the Eu^{2+} moments in the adjacent layers certainly accompanies modification of magnetic interactions with the Fe orbitals. It seems likely that this scenario may ascribe to a factor making the orbital pair-breaking effect insignificant.

To the best of our knowledge, there are only a few observations of field-induced superconductivity in URhGe [13], $Eu_xSn_{1-x}Mo_6S_8$ [14] and λ-$(BETS)_2FeCl_4$ [15]. URhGe is a special

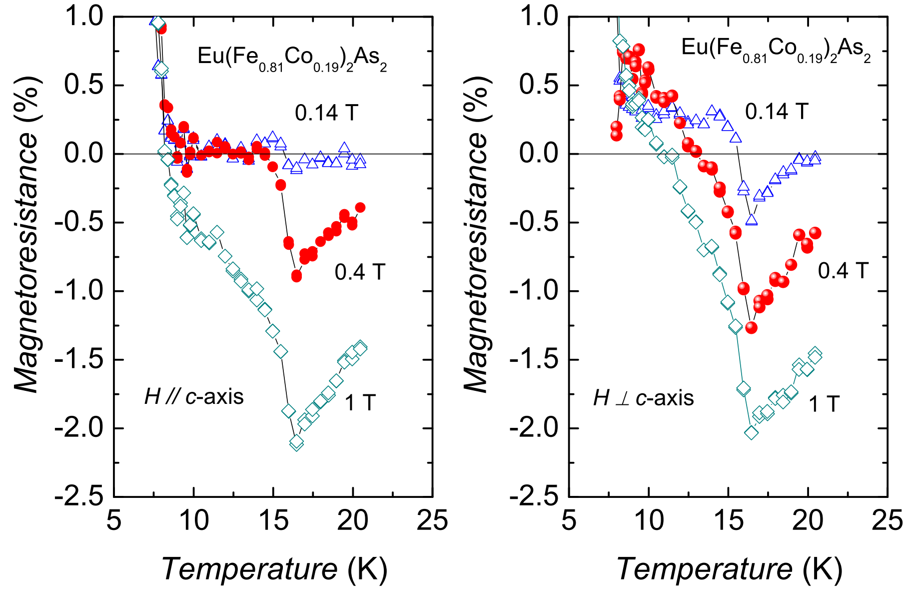

Figure 5: Temperature dependence of the magnetoresistance.

case, since the field-induced superconductivity may happen in neighborhood of a quantum transition under high magnetic field [13]. The behaviour of the second compound has been explained by the Jaccarino-Peter effect [16], i.e. a compensation between the external field and the internal field created by the the polarization of magnetic ions. For λ-$(BETS)_2FeCl_4$, besides possible Jaccarino-Peter compensation effect, the low dimensionality of the electronic system has been evoked for the interpretation [15]. In $Eu(Fe_{0.81}Co_{0.19})_2As_2$, the reason for inducing the superconductivity may be the following. In the strongly anisotropic systems, the orbital pair-breaking effect may be suppressed if the magnetic fields is applied parallel to the conductive direction [17]. As a result, since movement of the electrons perpendicular to the field is limited, the effect of the field is strongly suppressed, so is the orbital effect. However, even small component of the field/magnetisation parallel to the c direction may be sufficient to cause the orbital effect and break down the superconductivity completely. In the present case, the small component may result from canting of the Eu^{2+} moments toward the c axis in zero field. Furthermore, the magnetic field applied within ab planes polarizes Eu^{2+} ferromagnetically along the field direction, aligning the Eu^{2+} moments within the ab planes, and therefore the superconductivity can be restored. We may add that the comparison of

$Eu(Fe_{0.81}Co_{0.19})_2As_2$ with a non-magnetic superconducting $Ca(Fe_{0.96}Co_{0.04})_2As_2$ reference, adopting the same crystal structure and similar chemical stoichiometry but with different magnetic sublattices, [9] strongly supports our interpretation that the field-induced superconductivity in $Eu(Fe_{0.81}Co_{0.19})_2As_2$ is essentially associated with the interplay between magnetic interactions and orbital pair-breaking effect.

We have revealed that in the $Eu(Fe_{0.81}Co_{0.19})_2As_2$ single crystals external magnetic field applied perpendicular to the c axis leads to emergence of superconductivity. In order to explain the finding we have discussed several mechanisms, but the most plausible is that orbital pair-breaking effect can be abated by enhancement of the ferromagnetic interactions of the Eu^{2+} moments. We believe that the field tuning superconductivity opens new possibilities towards fabrication of field-controlled devices.

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[1] Marchand R. & Jeitschko W., Ternary lanthanoid-transition metal pnictides with $ThCr_2Si_2$-type structure, *J. Solid State Chem.* **24**, 351 - 357 (1978).

[2] Ren, Z. *et al.* Antiferromagnetic transition in $EuFe_2As_2$: A possible parent compound for superconductors, *Phys. Rev.* B **78**, 052501 (p4) (2008).

[3] Jeevan, H. S. *et al.*, Electrical resistivity and specific heat of single-crystalline $EuFe_2As_2$: A magnetic homologue of $SrFe_2As_2$, *Phys. Rev.* B 78, 052502 (p4) (2008).

[4] Wu, D. *et al.*, Effects of magnetic ordering on dynamical conductivity: Optical investigations of $EuFe_2As_2$ single crystals, *Phys. Rev.* B 79, 155103 (p10) (2009).

[5] Miclea, C. F., *et al.* Evidence for a reentrant superconducting state in $EuFe_2As_2$ under pressure, *Phys. Rev.* B 79, 212509 (p4) (2009).

[6] Terashima, T., *et al.,* $EuFe_2As_2$ under High Pressure: An antiferromagnetic bulk superconductor, *J. Phys. Soc. Jpn.* **78**, 083701 (p4) (2009).

[7] Zheng, Q. *et. al.*, Magnetism and superconductivity in single crystals $Eu_{1-x}Sr_xFe_{2-y}Co_yAs_2$, arXiv:0907.5547

[8] Matusiak M., Bukowski Z., Karpinski J., Doping dependence of the Nernst effect in $Eu(Fe_{1-x}Co_x)_2As_2$: Departure from Dirac-fermion physics, *Phys. Rev.* B **83**, 224505(p5) (2011).

[9] Supplementary information - available on request.

[10] Błachowski, A., *et al.*, Interplay between magnetism and superconductivity in $EuFe_{2-x}Co_xAs_2$ studied by ^{57}Fe and ^{151}Eu Mössbauer spectroscopy, *Phys. Rev.* B **84**, 174503 (p8) (2011).

[11] Werthamer, N. R., Helfand, E. & Hohenberg P. C., Temperature and purity dependence of the superconducting critical field, Hc2. III. Electron spin and spin-orbit effects, *Phys. Rev.* **147**, 295 - 302 (1966).

[12] Clogston, A. M., Upper limit for the critical Field in Hard Superconductors, *Phys. Rev. Lett.* **9**, 266 - 267 (1962).

[13] Lev F., Sheikin I., Grenier B. & Huxley A.D., Magnetic field-induced Superconductivity in the ferromagnet URhGe, *Science* **309**, 1343 - 1346 (92005).

[14] Meul, H. E. *et al.*, Observation of magnetic-field-induced superconductivity, *Phys. Rev. Lett.* **53**, 497-500 (1984).

[15] Uji, S. *et al.*, Magnetic-field-induced superconductivity in a two-dimensional organic conductor, *Nature* **410**, 908-910 (2001).

[16] Jaccarino, V. & Peter, M. Ultra-high-field superconductivity, *Phys. Rev. Lett.* **9**, 290 -292 (1962).

[17] Klemm, R. A., Luther A. & Beasley M. R., Theory of the upper critical field in layered superconductors, *Phys. Rev.* B **12**, 877 - 891 (1975).